INCORPORATING CO$_2$ SEQUESTRATION AND COALBED METHANE RECOVERY INTO HYDROGEN PRODUCTION FROM COAL - ECONOMICS AND ENVIRONMENTAL ASPECTS

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ABSTRACT

A hydrogen production process using pressure swing adsorption (PSA) for purification results in a concentrated CO$_2$ gas stream. In a typical natural gas steam reforming process this stream is used to fuel the reformer. However, because coal gasification takes place at high temperatures the synthesis gas contains very little CH$_4$ and other hydrocarbons, therefore, reforming is not required. An analysis was performed to examine hydrogen production via gasification of low sulfur western coal with CO$_2$ sequestration of the PSA off gas. This stream is then used to displace methane from unmineable coalbeds and the methane is utilized within the gasification-to-hydrogen system. Several processing schemes were evaluated: a reference case, a CO$_2$ sequestration only case, a maximum hydrogen production case, and a hydrogen/power coproduction case. The purpose of the analysis was to examine the technoeconomic feasibility, CO$_2$ emissions, and energy balance of these systems. This paper discusses the cases examined and presents the results of this study.

INTRODUCTION

A collaborative effort to study the feasibility of producing hydrogen from low Btu western coal with an emphasis on CO$_2$ sequestration and coal bed methane recovery was undertaken by National Renewable Energy Laboratory (NREL) and the Federal Energy Technology Center (FETC). Four cases which are outlined in Table 1 were studied in this analysis.

Table 1: Cases Examined

Case	Title	Description
1	reference case	coal gasification, shift, & H$_2$ purification
2	CO$_2$ sequestration only	reference case with CO$_2$ sequestration only added
3	maximum H$_2$ production	H$_2$ production via the syngas, CO$_2$ sequestration, & additional H$_2$ production via steam methane reforming of the coalbed methane
4	H$_2$/power coproduction	H$_2$ production via the syngas, CO$_2$ sequestration, & power production via the coalbed methane

SYSTEM DESCRIPTION

Because Wyodak coal is inexpensive to produce and is available in an abundant supply, it was selected as a suitable low-rank Western coal for this study. The coal is assumed to be mined from the Wyoming region, gasified to produce hydrogen, then the CO$_2$-rich off gas is injected into unmineable coal beds. Coal gasification is via the Destec gasifier which is a two-stage entrained, upflow gasifier that operates at an exit temperature of 1,038°C (1,900°F) and a pressure of 2,841 kPa (412 psia). The feed is a coal/water slurry and for hydrogen production, the gasifier is oxygen blown in order to minimize the amount of nitrogen in the syngas.

The synthesis gas leaving the gasifier contains entrained particles of char and ash. Particulate removal is performed through cyclone separators and ceramic candle type hot gas filters. The coal gas is primarily comprised of H$_2$, CO, CO$_2$, and H$_2$O and, since there is less than 0.1 mol% CH$_4$, reforming of the syngas is not necessary. However, in order to maximize hydrogen production, shift reactors are needed to convert the carbon monoxide to hydrogen. Because the syngas from the gasifier contains approximately 1,400 ppm of H$_2$S, the majority of the sulfur must be removed prior to shift conversion. Hot gas clean up (HGCU) followed by a ZnO bed is the most economical sulfur removal choice because it avoids cooling and reheating the syngas stream, in addition to avoiding condensing out the majority of the steam that is required for shift conversion. The transport desulfurizer technology from the Piñon Pine Project located near Reno, Nevada was used in the HGCU process step. This technology has an absorber/regenerator system where sulfur compounds

are absorbed on a zinc oxide based sorbent. When the sorbent is regenerated, SO_2 is captured and converted to sulfuric acid. Because the gasifier operates at a high temperature, a steam cycle was incorporated into the process design. Stepwise cooling of the synthesis gas produced steam that was used to generate electricity or to fulfill the plant steam requirements. Finally, hydrogen purification is done using a pressure swing adsorption unit.

The analysis assumes that two molecules of CO_2 were injected for every one molecule of CH_4 released from the coalbed (Gunter *et al*, 1996 and Hendriks, 1994). The off gas from the hydrogen purification unit containing primarily CO_2 (68 mol%; 93 wt%) was compressed from 2.6 MPa (372 psi) to a pressure of 8 MPa (1,160 psi). The analysis also assumed that new wells needed to be drilled and that they were connected by a CO_2 distribution system.

SCHEMES EXAMINED

In order to compare the economics as well as the overall CO_2 emissions from each schematic studied in this joint venture, a reference case was analyzed which included only the process steps associated with coal gasification, shift, and hydrogen purification (i.e., none of the steps associated with CO_2 sequestration or coalbed methane recovery were included in the reference case). Three other process schemes were examined in this study and compared to the reference case. Figure 1 depicts the reference case as well as the other three schemes (Note: The overall heat integration for each option is not shown.). The top portion of the figure shows the process steps that are the same for each schematic up to hydrogen purification while the operations inside the dashed boxes represent the steps that differ among the four options.

Scheme one represents the reference case (case 1). The PSA off gas is typically used to fuel the reformer in steam methane reforming plants but, due to the composition of the gasifier syngas, this scenario did not require a reformer. Therefore, the PSA off gas would be combusted, the heat would be used by another source (i.e., in producing steam), and the flue gas emitted to the atmosphere. The second scheme is the reference case with CO_2 sequestration only added to it and thus coalbed methane is not recovered (case 2). Scheme number three is maximum hydrogen production (case 3). The off gas is injected into the coal seam and a portion of the recovered methane is reformed to produce synthesis gas. This gas is then shifted and purified to produce more hydrogen. Part of the recovered coalbed methane is used to fuel the reformer. The fourth scheme is to produce hydrogen from the synthesis gas, to inject the CO_2-rich off gas into the coalbed, and to produce power from the recovered methane (case 4). For this scenario, power is produced using a natural gas turbine and steam cycle.

Figure 1: Systems Analyzed

These processing steps are the same for each schematic studied.
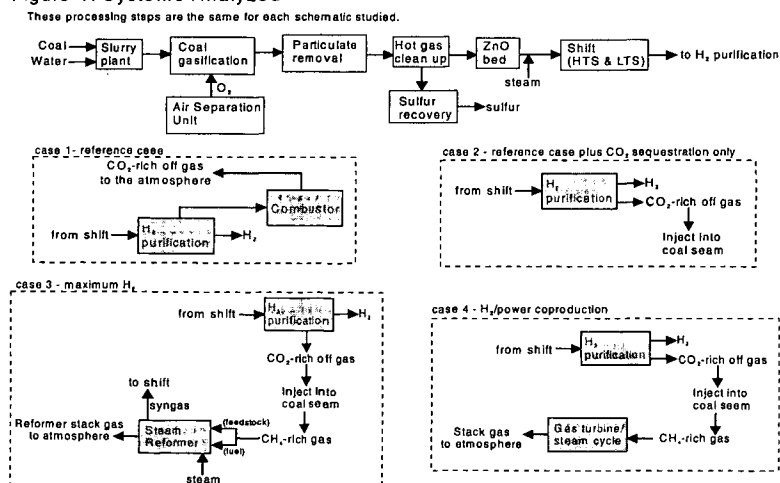
RESULTS - MATERIAL & ENERGY BALANCE

The material and energy balance for each case studied is given in Table 2. The coal feed rate is the same for each option and the resulting amount of hydrogen does not change except for the maximum hydrogen production case (case 3). Additionally, all of the cases examined, except maximum hydrogen production (case 3), produce some amount of power.

Table 2: Material & Energy Balances at 100% Capacity

Case	Coal (as received) (kg/hr)	Coalbed CH_4 (kg/hr)	H_2 (kg/hr)	Electricity required (MW)	Energy ratio
Reference	113,393	0	8,011	-12	0.83[1] 0.58[2]
CO_2 sequestration only	113,393	0	8,011	-4	0.57
Maximum H_2	113,393	47,366	18,739	3	0.67
H_2/power coproduction	113,393	36,419	8,011	-241	0.50

Energy ratio defined as (energy out of the system/energy into the system):

$$\frac{(H_2)(HHV_{H2}) + (STM_{ex})(\Delta H_{sh}) + e_{exp} + (off)(HHV_{off})}{(Coal_f)(HHV_{coal}) + (CH_4)(HHV_{CH4}) + e_{imp}}$$

H_2 = hydrogen produced (kg)
HHV_{H2} = higher heating value of hydrogen (GJ/kg)
STM_{ex} = steam produced which is sold (kg)
ΔH_{sh} = enthalpy difference between incoming water and steam produced which is sold (GJ)
e_{exp} = exported electricity (GJ equivalents)
off = off gas produced (kg) - reference case only
HHV_{off} = higher heating value of off gas (GJ/kg) - reference case only
$Coal_f$ = coal feed rate (kg)
HHV_{coal} = higher heating value of the coal (GJ/kg)
CH_4 = methane feed rate (kg)
HHV_{CH4} = higher heating value of the methane (GJ/kg)
e_{imp} = imported electricity (GJ equivalents)

[1]This is the energy ratio with an off gas energy credit.
[2]This would be the energy ratio if there were no off gas energy credit.

RESULTS - CO_2 BALANCE

To adequately determine the overall effect of CO_2 for each option studied, the CO_2 balance must incorporate CO_2 emissions in addition to those emitted from the process itself. For example, each case produces electricity, except for the maximum hydrogen production case (case 3), and for these cases (cases 1, 2, and 4) a CO_2 emissions credit must be taken for displacing electricity from the grid. Because the maximum hydrogen production case (case 3) requires some grid electricity, the system must be debited (rather than credited) with CO_2 emissions equivalent to the plant's net electricity requirement. Additionally, for the two options that recover coalbed methane (case 3 and 4), each of those systems must be credited with CO_2 emissions that are avoided from natural gas production and distribution via today's normal routes of gas and oil wells. Table 3 summarizes the CO_2 emissions for each of the cases examined.

Table 3: CO_2 Balance at 100% Capacity

Case	Overall CO_2 to atm (kg/hr)	Avoided electricity CO_2 (kg/hr)	Avoided natural gas CO_2 (kg/hr)	Electricity CO_2 (kg/hr)	Process CO_2 (kg/hr)
Reference	195,707	-10,037	N/A	N/A	205,744
CO_2 sequestration only	-3,667	-3,667	N/A	N/A	0
Maximum H_2	65,985	N/A	-12,694	2,619	43,070
H_2/power coproduction	-109,065	-200,575	-9,760	N/A	101,270

Process CO_2 defined as:
Reference = flue gas resulting from combusting CO_2-rich PSA off gas
CO_2 sequestration only = none
Maximum H_2 = CO_2 in the reformer flue gas
H_2/power coproduction = CO_2 in the natural gas combined cycle stack gas

For the reference case (case 1), the CO_2 emissions are primarily a result of the hydrogen production process. The overall CO_2 emissions for the CO_2 sequestration only case (case 2) are actually slightly negative instead of zero because of the CO_2 credit for the displaced grid electricity. The hydrogen/power coproduction case (case 4) also results in a negative amount of CO_2 emissions. This is due to the large credit in CO_2 emissions from displacing a significant quantity of grid electricity. In this analysis, grid electricity was assumed to be the generation mix of the mid-continental United States, which according to the National Electric Reliability Council, is composed of 64.7% coal and

coal-fired power plants generate large quantities of CO_2. There are still a large amount of process emissions from this system but these are overshadowed by the avoided CO_2 emissions. Even though the maximum hydrogen case (case 3) sequesters CO_2, some CO_2 is generated when the off gas is burned in the reformer. Also, some electricity is required for this case which results in additional CO_2 emissions. However, overall it is not correct to compare the emissions on a per system or a per amount of hydrogen produced basis because many of these cases generate power (refer to Table 2) and all of the cases produced energy in the form of steam. Additionally, for two of the cases (case 3 and 4), the additional hydrogen or power is produced from coalbed methane and the energy content of this feedstock must be taken into consideration. To correctly compare each system, they must be examined on an energy wide basis. If the CO_2 emissions were examined per the amount of hydrogen produced from each system then the results would be misleading. Therefore, the CO_2 emissions were divided by the energy ratio of the system and the results can be seen in Figure 2. For comparison, the CO_2 emissions were also plotted assuming that no CO_2 credits or debits were taken for grid electricity and natural gas production and distribution. It is evident that the only case that is greatly affected by this, and would most definitely be misrepresented, is the hydrogen/power coproduction case (case 4). Hydrogen/power coproduction (case 4) and CO_2 sequestration only (case 2) are the only cases that result in a negative amount of CO_2 emissions. However, the maximum hydrogen production case (case 3) does emit significantly less CO_2 than the reference case (case 1).

Figure 2: CO_2 Emissions per Energy Ratio @ 90% Capacity Factor
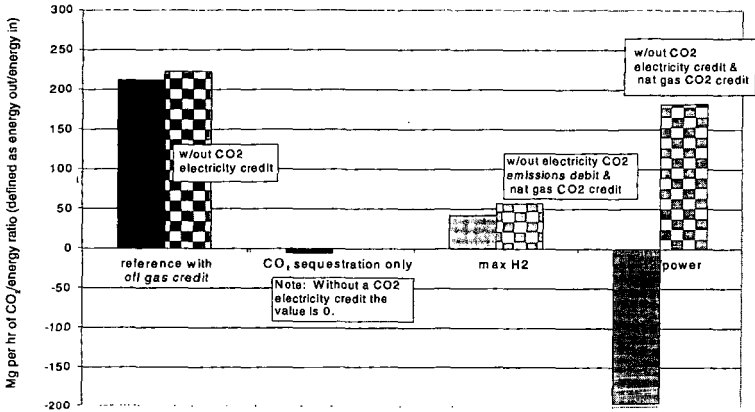
RESULTS - COST OF HYDROGEN

The plant gate hydrogen selling price was determined using a cash flow analysis with an after-tax internal rate of return (IRR) of 15%. However, because the hydrogen production plant must be located at the coal mining site in order to sequester CO_2 into unmineable coalbeds, it is not likely that the consumer will be "over the fence". Therefore, the cost to store and transport the hydrogen must be added to the plant gate cost in order to determine the delivered cost of the hydrogen. For this study, because the hydrogen plant is assumed to be sited far from any users, two likely storage and transportation options were examined: (1) bulk delivery for a distance of 1,610 km (one way) and (2) pipeline delivery with 3 km to nearest infrastructure; no storage; an additional 1,610 km pipeline distance for delivery to end user for which the cost is shared by 5 companies. Table 4 gives the plant gate cost as well as the delivered cost for each of the cases examined in this study. The cheapest storage method for the bulk delivery option was determined to be liquid/rail based on previous work at NREL (Amos, 1998). The maximum hydrogen production and hydrogen/power coproduction cases (case 3 and 4) were found to be more economical than the reference case (case 1). Pipeline delivery added \$4.67/GJ to the plant gate cost and bulk delivery added \$8.78/GJ.

Table 4: Cost of Hydrogen

Case	Plant gate hydrogen selling price ($/GJ)	Delivered cost of hydrogen ($/GJ)	
		Bulk delivery (1,610 km - liquid/rail)	Pipeline delivery (1,610 km - shared)
Reference	17.98	26.76	22.65
CO_2 sequestration only	18.72	27.50	23.39
Maximum hydrogen	8.34	17.12	13.01
H_2/power coproduction	13.92	22.70	18.59

RESULTS - CARBON TAX

By comparing the hydrogen selling price of the reference case (case 1) with that of the CO_2 sequestration only case (case 2), a carbon tax that would represent a break-even point was calculated. The hydrogen selling price for the CO_2 sequestration only case (case 2) would be reduced to $17.98/GJ, the reference (case 1) with off gas credit case cost, if a carbon tax of $13.4/tonne of carbon was mandated. Additionally, to examine the affect of a higher tax, a carbon tax of $50/tonne of carbon was applied to the analysis. Figure 3 shows the plant gate selling price (denoted by the checkered sections), the cost of hydrogen for the baseline pipeline delivery option, and how the two carbon tax values affect the delivered cost of hydrogen for each of the four cases examined in this study. A carbon tax has the greatest effect on the hydrogen/power coproduction case (case 4). A $50/tonne of carbon tax brings the delivered price of hydrogen below the plant gate selling price with the delivered cost being reduced from $18.6/GJ to $13.3/GJ. The delivered cost of hydrogen from the CO_2 sequestration only case (case 2) is reduced by $2.8/GJ while a carbon tax has a small effect of the maximum hydrogen case (case 3) with a reduction of about $1/GJ.

Figure 3: Delivered Cost of Hydrogen (Pipeline -1,610 km) with a Carbon Tax
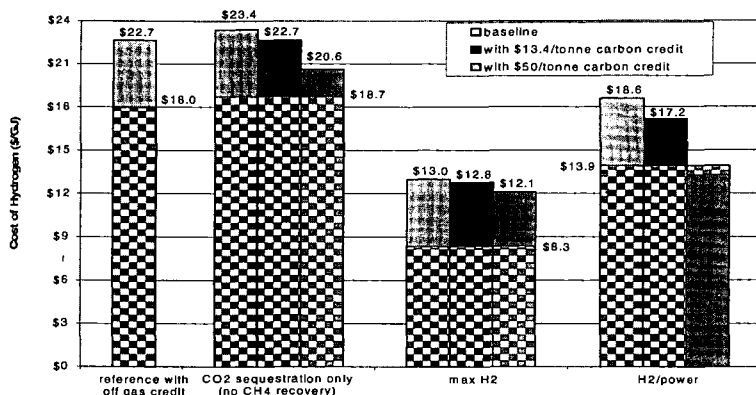
CONCLUSIONS

Four process schemes were evaluated in this coal gasification, hydrogen production study. The economics favor sequestering CO_2, recovering coalbed methane, and making hydrogen or power (case 3 and 4). However, due to the CO_2 emissions generated from the steam methane reformer, additional hydrogen production via natural gas is not necessarily the most environmentally friendly option from a CO_2 standpoint (case 3). Coal fired power plants emit large quantities of CO_2, therefore optimizing hydrogen production with electricity generation, as in case 4, is a means of lowering the CO_2 emissions from power generation in the U.S. Because of the high temperatures, coal gasification to hydrogen production does not require a steam methane reforming step, and adding CO_2 sequestration only (case 2), results in almost no CO_2 being emitted to the atmosphere for a minimal cost. Mandating a carbon tax would make sequestering the CO_2 economically viable. However, for all of the cases examined in the analysis it should be noted that there is much debate about the fate of the sequestered CO_2 and its long term environmental effects.

ACKNOWLEDGEMENTS

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